

INEEL/EXT-2000-00959

September 2000



**Idaho National Engineering and  
Environmental Laboratory  
Site Report on the Production  
and Use of Recycled Uranium**

*L. C. Lewis  
D.C. Barg  
C. L. Bendixsen  
J. P. Henscheid  
D.R. Wenzel  
B. L. Denning*



**IDAHO NATIONAL ENGINEERING AND  
ENVIRONMENTAL LABORATORY  
SITE REPORT ON THE PRODUCTION  
AND USE OF RECYCLED URANIUM**

**L. C. Lewis  
D. C. Barg  
C. L. Bendixsen  
J. P. Henscheid  
D. R. Wenzel  
B. L. Denning**

**Published September 2000**

**Idaho National Engineering and Environmental Laboratory  
Idaho Falls, Idaho 83415**

**Prepared for the  
U.S. Department of Energy  
Assistant Secretary for  
Environmental Management  
Under DOE Idaho Operations Office  
Contract DE-AC07-99ID13727**

## Site Team Approval Sheet



**L. C. Lewis, Site Lead  
Process Chemist**

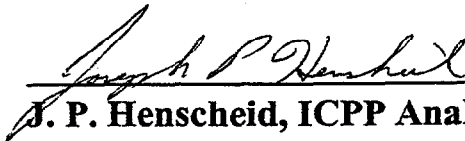


6/26/00

**D. C. Barg, SMC Lead  
SMC Health Physics**



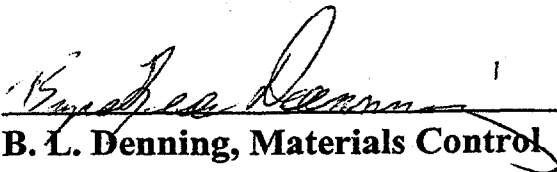
**C. L. Bendixsen, ICPP Process Engineering**



**J. P. Henscheid, ICPP Analytical Chemistry**



**D. R. Wenzel, ICPP Health Physics**



**B. L. Denning, Materials Control  
and Accountability**

## Executive Summary

Recent allegations regarding radiation exposure to radionuclides present in recycled uranium sent to the gaseous diffusion plants prompted the Department of Energy to undertake a system-wide study of recycled uranium. Of particular interest, were the flowpaths from site to site, operations and facilities in which exposure to plutonium, neptunium and technetium could occur, and to the workers that could receive a significant radiation dose from handling recycled uranium.

The Idaho site report is primarily concerned with two locations at the Idaho site. Recycled uranium was produced at the Idaho Chemical Processing Plant where highly enriched uranium was recovered from spent fuel. The other facility is the Specific Manufacturing Facility (SMC) where recycled, depleted uranium is manufactured into shapes for use by their customer.

The Specific Manufacturing Capability (SMC) is located in the Test Area North, which was originally built in the late 1950's to develop the nuclear aircraft. This development project was terminated and the SMC complex was later installed in the nuclear aircraft project building. SMC's current mission is the fabrication of components from depleted uranium for government purposes.

The SMC is a manufacturing facility that uses depleted uranium metal as a raw material that is then rolled and cut into shapes. There are no chemical processes that might concentrate any of the radioactive contaminant species. Recyclable depleted uranium from the SMC facility is sent to a private metallurgical facility for recasting. Analyses on the recast billets indicate that there is no change in the concentrations of transuranics as a result of the recasting process.

The Idaho Chemical Processing Plant is located in south-eastern Idaho at the Idaho National Engineering and Environmental Laboratory (INEEL). The facility was built to recover high-enriched uranium from spent nuclear fuel from test reactors. The facility processed diverse types of fuel which required uniquely different fuel dissolution processes. The dissolved fuel was passed through three cycles of solvent extraction which resulted in a concentrated uranyl nitrate product. For the first half of the operating period, the uranium was shipped as the concentrated solution. For the second half of the operating period the uranium solution was thermally converted granular, uranium trioxide solids.

Approximately 85% of the uranium product was shipped to the Y-12 facility at Oak Ridge. Most of the rest was shipped to the Portsmouth Gaseous Diffusion Plant. Small quantities were shipped to Rocky Flats, Pacific Northwest National Laboratory, and to Los Alamos for their use in criticality experiments.

Shipments from ICPP were begun in 1953 and continued until 1998. During this time period there was 32,005 tonnes of high enriched uranium product produced. In addition, there was approximately 20 Kg of material received at ICPP from Y-12 which was a denitrated uranium trioxide which was to be used as the start up bed for denitrating the product. A second shipment

was received from Pacific Northwest National Laboratory at the conclusion of their criticality experiments. The material that was sent back was approximately one-half of the 47 Kgs of uranium that was sent to them in 1978. There were three shipments of uranium from the processing of the stainless steel clad EBR-II fuel consisting of a total of 4.08 metric tonnes of uranium at an enrichment of 50%. There was also 219.10 Kgs sent to Rocky Flats in 1955 and there was 167.61 Kgs sent to Los Alamos in 1984. There is 1.770 tonnes of uranium currently in storage at ICPP. Everything else was shipped to Y-12.

Throughout the history of the ICPP, the uranium product was monitored for its transuranic alpha content, beta content and occasionally for its gamma content. The alpha content was consistently below the informal and formal specification. In the early years the beta ratio was greater than the specification but this was also reduced to a level below the specification limits. The beta emitting contaminant was primarily ruthenium because it was not very effectively removed by the hexone extraction cycles. When the tributyl phosphate cycle was introduced the ruthenium concentration decreased. Uranium-236 and uranium-234 were also significant contaminants in the ICPP product. Uranium-236 was produced by activation of the uranium while it was in the reactor, while uranium-234 was preferentially enriched in the gaseous diffusion plants; and neither uranium isotope could be removed by chemical processing. Technetium-99 was not measured in the uranium product because it was not considered to be a problem during all the years of processing. Its concentration was believed to be insignificant compared to ruthenium.

Currently ICPP has in its recycled uranium product inventory, 1.770 MTU of high enriched uranium trioxide. Most of this material contains a high concentration of U-236 which can result in significant gamma fields when secular equilibrium is approached.

Worker exposure occurred throughout the operating history of the ICPP as the result of normal operations, maintenance activities, analytical chemistry activities, and health physics activities. In the early years personnel were pushed close to the annual or quarterly limits. From the mid 1970s on, workers were closely monitored to make certain that they did not exceed 3 rem per year. The facilities in which exposures took place included all of the facilities where irradiated material was handled or stored. These facilities included CPP-603, CPP-601, CPP-602, CPP-627, CPP-640, CPP-684, CPP-604, CPP-630, CPP-633, CPP-666, and CPP-659. The facilities were the primary fuel processing, waste processing, maintenance, analytical chemistry, and fuel storage facilities. All of these facilities contributed to worker exposure because the ICPP facility was a direct maintenance facility.

The dose reconstruction project has evaluated worker exposure and exposure to the public as the result of normal operations and accidents that occurred at the INEEL. As a result of these studies, the maximum effective dose equivalent from site activities did not exceed seventeen percent of the natural background in Eastern Idaho. There was no year in which the radiation dose to the public exceeded the applicable limits for that year. Worker exposure to recycled uranium was minimized by engineering features that reduced the possibility of direct exposure.

The SMC facility only worked with depleted uranium metal. It received only one lot, and all of its processing activities have been with that lot of material. Metallic waste has been sent to a private recasting company. The quantities of transuranics and technetium have been below the *de minimis* levels, and SMC performs no operations that would result in concentrating or release of any of the contaminants. There have been no releases of this material to the environment from the SMC site. No uranium attributable to SMC operations has been found outside the SMC facility fence.





## Table of Contents

Title Page	i
Executive Summary	iii
Table of Contents	vi
List of Table	viii
List of Figures	ix
Acronyms	x
Introduction to the INEEL Report	1
1.0 Idaho National Engineering and Environmental Laboratory Recycled Uranium Mass Balance Project	2
1.1 Project Overview	2
1.2 Purpose and Scope	3
1.3 Project Implementation Strategies	4
2.0 Site Historical Overview	5
2.1 Idaho Chemical Processing Plant Location	5
2.2 Key Uranium Processing Facilities	5
2.2.1 Idaho Chemical Processing Plant	8
2.2.1.1 Plant Description	8
2.2.1.2 Material Flowsheet	14
2.2.1.3 Feed Specifications	15
2.2.1.4 Product Specifications	15
2.2.1.5 Operating History	15
2.2.1.6 Current Status	15
2.3 Activity Summaries	15
2.3.1 Bottling Liquid Product	18
2.3.2 Packaging Solid Product	18
2.3.3 Analysis of Liquid Product	18
2.3.4 Operating the Denitrator	18
2.3.5 Maintenance on the Denitrator	18
2.3.6 Health Physics Surveillance During Denitrator Operation	18
2.3.7 Health Physics Monitoring During Liquid Product Bottling	19
2.4 Work Force Exposure	19
2.5 Environmental Releases	24
3.0 Recycled Uranium Mass Flow	26
3.1 Uranium Recycle Description	26
3.2 Uranium Receipts	26
3.3 Uranium Shipments	26
4.0 Constituents in Recycled Uranium	36
4.1 Analytical Laboratories	36
4.1.1 Analytical Methods	36
4.1.2 Analytical Methods	36

## Table of Contents (continued)

4.1.3	Processing Issues	36
4.1.4	Quality Assurance	36
4.2	Neptunium, Plutonium, and Technetium in ICPP Uranium Product as Estimated by ORIGEN2 Calculations	38
4.3	Analytical Results for Plutonium	46
4.3.1	Plutonium Specification	
4.3.2	Impurity Concentrations for Plutonium in Materials Shipped	46
4.4	Analytical Results for Neptunium in Uranium Materials Shipped	47
4.4.1	Neptunium Specifications Uranium Materials Shipped	47
4.4.2	Impurity Concentration for Neptunium in Recycled Uranium Shipped	47
4.5	Analytical Results for Technetium in Uranium Materials Shipped	47
4.5.1	Technetium Specification in Recycled Uranium	47
4.5.2	Impurity Concentration for Technetium in Uranium Materials Shipped	47
4.6	Analytical Results for Material Received	47
4.7	Discussion of Other Constituents	47
5.0	Mass Balance Activities	51
5.1	Annual Mass Balance of Recycled Uranium	51
5.2	Annual Mass Balance for Plutonium	51
5.3	Annual mass Balance for Neptunium	51
5.4	Annual Mass Balance for Technetium-99	51
5.5	Annual Mass Balance for Other Constituents	55
5.6	Potential for Worker Exposure from Recycled Uranium	55
5.7	Potential for Environmental Contamination from Recycled Uranium	55
6.0	Results and Conclusions	55
6.1	Explanation of mass Flow Paths and Contaminant Levels	55
6.2	Identification of Processes or Areas of Concern for Worker Exposure	56
6.3	Identification of Processes or Areas of Concern for Environmental Impact	56
6.4	Discussion of Data Sources and Confidence Levels	56
6.5	Conclusions	58
7.0	References	59
	Appendix	60

## Tables

Table I	ICPP Activity Chart	17
Table II	Lung Clearance Classes Used to Determine the Relative Hazard from Various Isotopes	22
Table III	Comparative Risk and Effective Dose Equivalent for Isotopes in the Product from Processing at ICPP	23
Table IV	Shipments of Final Product	28
Table V	Fuel Processed at ICPP	29
Table VI	ORIGEN2 Results in Terms of Grams/100grams of Uranium	40
Table VII	ORIGEN Result in Terms of Ci/gU	41
Table VIII	Comparison of Pu/U Mass Ratios from Measured Decontamination Factors and Alpha Ratios	44
Table IX	Contaminants in ICPP Product. Based on ORIGEN2 Code Calculations and DFs from ICPP Process Data	45
Table X	Uranium-236 Content of ICPP Fuels	48
Table XI	Uranium-236 Quantities Sent to Receiving Sites	49
Table XII	Concentration of Contaminants in ICPP Product	49
Table XIII	Contaminants in ICPP Product	50
Table XIV	Material Shipped from ICPP	50
Table XV	Recycled Uranium Shipment	52
Table XVI	Ranges of Contaminants	58

## List of Figures

Figure 1	The Idaho Chemical Processing Plant as it Exists Today	6
Figure 2	Historical Time Line of Important Events at Idaho Chemical Processing Plant	7
Figure 3	Flowsheet of Processes used at ICPP	9

## Appendix Figures

Figure A1	Simplified Chemical Flowsheet	
Figure A2	Processing Flowsheet for Dissolution of Aluminum Fuels and First Cycle Solvent System Processing of the Aluminum Dissolver Product	
Figure A3	Zirconium Processing Flowsheet for Campaigns 33 and 35	
Figure A4	Campaign 37 Electrolytic Dissolver Flowsheet for Processing Borax V Type Fuels (0.4 g SS dissolved/amp - hr)	
Figure A5	Campaign 37 Flowsheet for Processing Dissolver Product Through The First Cycle Extraction System with No Raffinate Recycle	
Figure A6	Coprocessing Dissolver Flowsheet for Campaign 30: PWR-ATR Fuels	
Figure A7	Flowsheet for Burnig of ROVER Fuel	
Figure A8	Flowsheet for Dissolution of ROVER Ash	
Figure A9	Campaign 37 Second and Third Cycle Extraction Flowsheet for High Uranium Concentration Feed	
Figure A10	Campaign 37 Denitration Flowsheet for concentrated $\text{UO}_2(\text{NO}_3)_2$ Solutions	

## Acronyms

AEC	Atomic Energy Commission
ALARA	As Low As Reasonably Achievable
AMAD	Activity Median Aerodynamic Diameter
BBWI	Bechtel BWXT Idaho
CEDE	Committed Effective Dose Equivalent
CPM	Continuous Processing Modification
DF	Decontamination Factor
DOE	Department of Energy
DPM (dpm)	Disintegration Per Minute
DPS (dps)	Disintegrations Per Second
DU	Depleted Uranium
EBR-I	Experimental Breeder Reactor I
EBR-II	Experimental Breeder Reactor II
FAST	Fluorinel and Storage Facility
FDP	Fluorinel Dissolution Process
FECF	Fuel Element Cutting Facility
HEU	High Enriched Uranium
ICPP	Idaho Chemical Processing Plant
IDMS	Isotope Dilution Mass Spectrometry
ISF	Irradiated Fuel Storage Facility
INEEL	Idaho National Engineering and Environmental Laboratory
MTR	Materials Testing Reactor
NBS	National Bureau of Standards
NIST	National Institute of Science and Technology
NP	Neutron Producing
NWCF	New Waste Calcination Facility
ORIGEN	Oak Ridge Isotope Generation and Depletion
PGDP	Portsmouth Gaseous Diffusion Plant
RAF	Remote Analytical Facility
RAL	Remote Analytical Laboratory
RALA	Radioactive Lanthanum
ROVER	Nuclear Rocket Program
SMC	Special Manufacture Capability
TAN	Test Area North
TRU	Transuranic
WCF	Waste Calcination Facility
Y-12	Weapons Plant at Oak Ridge, TN

